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THE LISA-1 AND TRIO IN-PILE TESTS

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I. INTRODUCTION

The in-pile tests TRIO[1] and LISA-1[2] involve measurement of in-situ tritium release as a funtion of time, temperature and sweep gas conditions. These in-situ tritium recovery experiments are similar in concept to other in-pile tests such as the VOM series,[3,4] Exotic,[5,6] and the LILA series.[7,8] TRIO used a single capsule with lithium aluminate. The results from TRIO have been compiled, evaluated and reported.[1] The LISA-1 experiment had six test capsules: one lithium aluminate, one lithium orthosilicate (Li_4SiO_4), and four lithium metasilicate (Li_2SiO_3). A previous report gives a description of the experimental setup, experimental observations, and preliminary results.[2] Presented herein is a more detailed evaluation of the LISA-1 experimental results for the three breeder materials. The results from LISA are then compared to those of TRIO.

II. EXPERIMENTAL DESCRIPTION

The setup for the experiments is summarized herein. Both LISA and TRIO involve neutron irradiation of specimens of solid breeders (Table 1) in capsules swept with a purge gas used for the purpose of in-situ tritium recovery. The sweep gas in each line passes through a set of water bubblers that collect tritiated water, a molecular seive dryer, then through the ion chamber where tritium activity is continuously measured, and finally through a CuO bed with a set of water bubblers. The tritium collected in both sets of bubblers is analysed by scintillation counting.

Table 1. Test Specimens

Sample	Matl.	Grain, r(μm)	% t.d.	mass (g)	Li-6 %	nv(th)/ E14
TRIO	$LiAlO_2$	0.1	65	42.9	0.55	1.1
LISA						
P1	Li_2SiO_3	~100	86	4.4	7.4	0.13
P3	Li_2SiO_3	~100	93	8.6	7.4	0.13
P5	Li_2SiO_3	~100	86	8.5	7.4	0.13
P6	Li_4SiO_4	~100*	95	8.4	7.4	0.13
P4	Li_2SiO_3	~100	86	8.3	7.4	0.13
P2	$LiAlO_2$	0.19	78	7.7	7.4	0.13

*Sample has almost no open porosity-effective "r" may be pellet, i.e., 4 mm.

III. LITHIUM ALUMINATE

The data on tritium release rates, in particular the dynamic response of the system to step changes in temperature or neutron flux, provides information that relates directly to tritium transport processes, notably intra-granular diffusion. Models have been developed based on analytical solutions to the diffusion equation.[1,4,8] The data treatment herein is essentially the same as Kurasawa, et al.[4]

An example comparing the observed tritium release curve with the diffusional calculations is shown in Fig. 1 for run C-2, a run with H_2 added to the sweep gas with temperature going from 596 to 548 C. The theoretical and experimental curves match well at longer times but are quite different at short time (1 hr or less). This behavior is consistent with the interpretation that bulk diffusion is generally the dominant mechanism but that there is some evidence for surface effects at very short times. This general behavior is observed consistently for lithium aluminate in the LISA-1 experiment. The time constants calculated from

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dynamic analysis of the response of the lithium aluminate sample for all the runs are given in Table 2. The results for pure He sweep gas are in a similar range with the results when H_2 was added, but the results are more consistent when H_2 is added. It is likely that surface effects are reduced when H_2 is added.

The results for both transient and steady-state analysis were used to calculate diffusivities at each temperature (Fig. 2). These values are compared to curves of diffusivities from TRIO and the VOM-22H experiment.[4] The values from both transient analysis and steady-state analysis are very close to those from VOM-22H and about a factor of 10 higher than TRIO. Note that the VOM-22H experiment has much larger (10 μm) grains. Differences of within a factor of ten for diffusivity measurements are considered to be in very good agreement. These results provide strong evidence that a single mechanism is controlling the tritium release, and that this mechanism is intragranular diffusion.

IV. LITHIUM ORTHOSILICATE

Dynamic analysis was also performed for the lithium orthosilicate. As an example, the calculated and observed curves for run B-7 (temperature 455-->508 C, 0.1 % H_2 in sweep gas) are shown in Fig. 3. The agreement of the two curves is quite good and only a small difference is seen at very short times. This difference may represent the lag times inherent in the experiment arising from a number of factors including: non-instantaneous temperature change, mixing times in the measuring system, and instrumental time constants. A comparison with the aluminate suggests that this sample (when H_2 is added to the sweep gas) has better agreement with pure diffusion at very short times. This might be expected, owing to the much larger grain size of this sample. The time constants for all the runs evaluated are given in Table 3.

Table 2. Time Constants from Dynamic Analysis - Sample P2

Run No.	T_2 C	Sweep Gas Flow	Sweep Gas Comp.	nV/10	τ_2, hr	T_1	τ_1 hr
A-2	540	1.0	He	1.300	4.9	594	2.3
A-3	593	"	"	1.306	3.75	540	10.4
A-4	540	"	"	1.229	7.5	593	3.8
A-5	500	"	"	1.230	18.5	540	4.6
A-6	542	"	"	1.241	8.0	500	29.2
A-7	594	"	"	1.242	4.2	542	16.8
B-4	504	"	+H2	1.282	31.	594	8.8
B-6	453	0.7	"	0.980	71.	504	23.7
B-7	505	1.0	"	1.290	10.	453	30.
B-8	595	"	"	1.305	1.2	505	15.
C-1	596	"	"	1.310	1.5		
C-2	548	"	"	1.295	1.85	596	0.9
C-3	548	"	"	2.173	2.		
C-4	548	"	"	1.300	2.5-3.5		
C-5	598	"	"	1.291	0.85	548	4.7
C-6	628	"	"	1.286	0.97	598	1.21
C-7	598	"	"	1.267	0.75	628	0.76

The time constants determined from steady-state and from dynamic analysis are compared in Fig. 4. It is evident that there is good agreement between dynamic and steady-state measurement. The runs with H_2 added to the sweep gas appear to fall on a single straight line. When H_2 is not added, the data shows longer time constants and has more scatter. This means that with H_2 added, the behavior is diffusion-like. By contrast, without H_2 added, the release is more complex. The values for diffusivity are not plotted because of uncertainty in the path length r. This may be represented by a value as small as the grain size or as large as the pellet size. More experiments with different textures will resolve this question.

V. LITHIUM METASILICATE

The metasilicate samples did not clearly have a good steady-state line. The release seemed to level off at a different asymptote at each temperature, with lower values for the asymptote at lower temperatures. This behavior is as if a certain fraction of the tritium is being trapped, and this fraction is higher at lower temperatures. Since steady-state was not reached for these samples, inventory analysis was not performed. However transient analysis was performed in an effort to determine time

Table 3. Time Constants from Dynamic Analysis - Sample P6

Run No.	T_2 C	Sweep Gas	n. Flux E13nv	τ_2 hrs	T_1,C	τ_1, hrs
A-1	604	1.0 He	1.297	3.50		
A-2	547	" "	1.300	3.75	604	2.04
A-3	603	" "	1.306	4.00	547	10.72
A-4	548	" "	1.229	8.80	603	5.58
A-5	508	" "	1.230	16.75	548	7.97
A-6	549	" "	1.241	8.0	508	23.0
A-7	606	" "	1.242	6.6	549	19.8
B-4	507	" +H2	1.282	9.5	602	5.57
B-6	455	0.7 "	0.980	11.5	507	5.47
B-7	508	1.0 "	1.290	4.0	455	9.5
B-8	600	" "	1.305	1.2	508	4.15
C-1	602	" "	1.310	0.88	602	1.24
C-2	555	" "	1.295	1.63		
C-3	555	" "	2.173	3.3		
C-4	555	" "	1.300	1.9-3.4		
C-5	603	" "	1.291	1.02	555	1.83
C-6	635	" "	1.286	1.10	603	1.26
C-7	604	" "	1.267	0.65	635	0.49

constants. The time constants are about 10 hours, at 600°C which translates to a diffusivity of 0.7 E-10 cm^2/s. The scatter is very large, however, so this value is only an estimate of the apparent diffusivity. There is no noticible effect of adding hydrogen to the sweep gas, in contrast to the aluminate and the orthosilicate.

VI. CONCLUSIONS

Although evaluation of the data from LISA-1 is not complete, certain conclusion and obervations can be made for the three breeder materials:

$LiAlO_2$ - Reasonably good agreement between TRIO and LISA-1 is evident with small-grained material. In addition, both show good agreement in apparent diffusivity with results of VOM-22H, which has larger grains and which spans the higher temperature range. These results provide evidence that intragranular diffusion is a dominant transport mechanism. At the same time, perhaps owing to the small grain size in LISA-1 there is some evidence of other factors, particularly at very short times.

Li_4SiO_4 - When hydrogen is added to the sweep gas, this material shows the best release, especially at low temperatures. Also, with H_2 added, the release appears to be diffusion-like and there is not evidence of other processes. The time constants and temperature dependance thereof for this polycrystalline material are in very good agreement with the data of Ihle[9] as was previously reported.[2] However, at the present time, there remains some uncertainty as to whether the true diffusion path length is the grain radius. Thus, more work is needed to determine whether these results in fact represent intragranular diffusion.

When no hydrogen is added to the sweep gas, the release behavior is more complex. It appears that diffusion is still important, but at least one other mechanism is occurring.

Li_2SiO_3 - The tritium release from these samples is complex and cannot be represented by single mechanism. Evaluation of time dependant behavior suggests that diffusion may be a contributing mechanism and that the diffusivity values are the order of 10^{-10} cm^2/s at 600 C.

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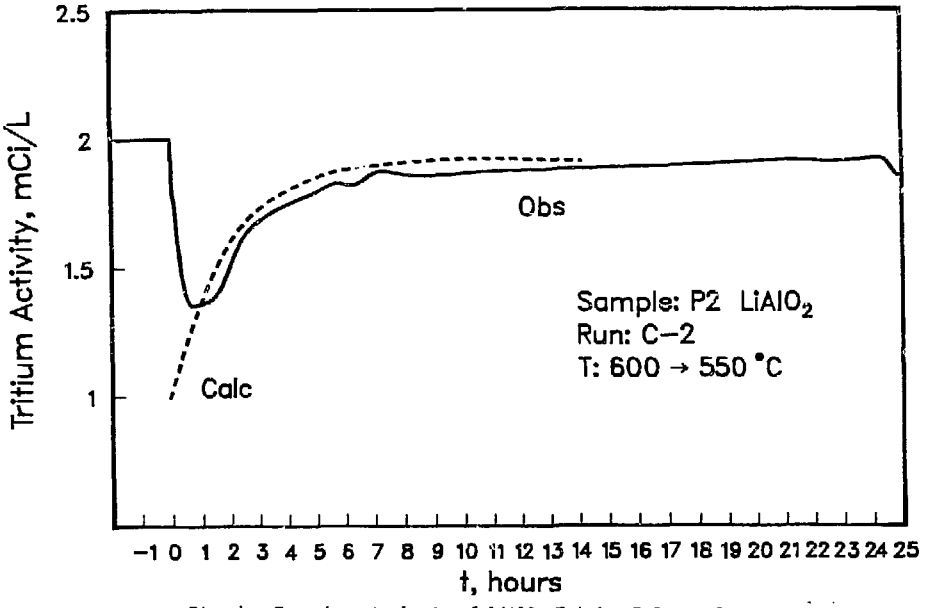

Fig. 1. Transient Analysis of $LiAlO_2$ Tritium Release Curve

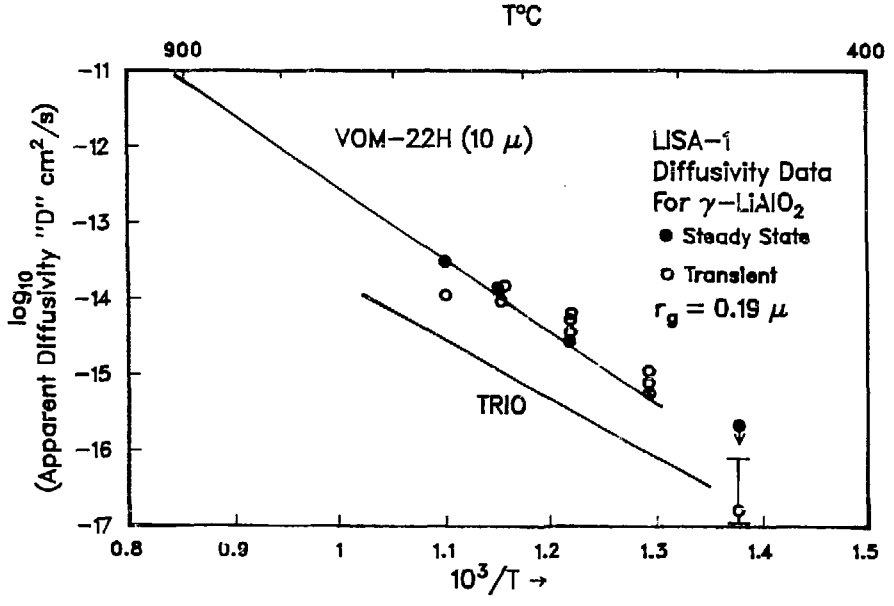

Fig. 2. Calculated Diffusivity Value from LISA-1 are Compared to Other In-Pile Tests

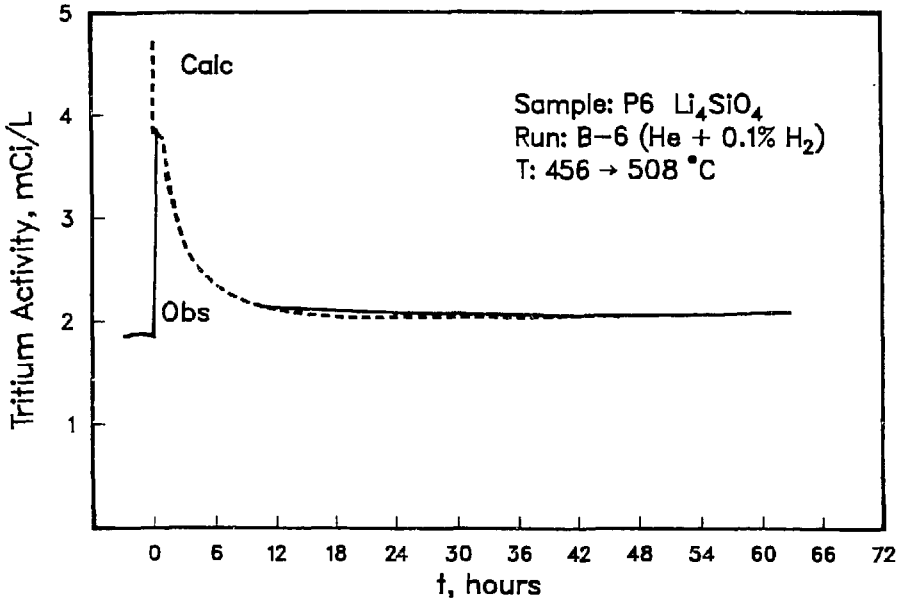

Fig. 3 Transient Analysis of Li_4SiO_4 Tritium Release Curve.

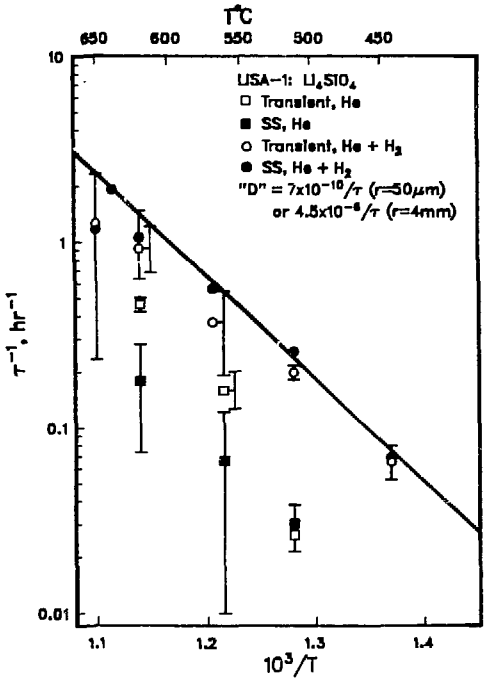

Fig. 4. Summary of Diffusion Results for Li_4SiO_4. Transient and Steady State (SS) Analysis are in Agreement.